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Oscillating magneto- and electrocaloric effects on bilayer graphenes

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ABSTRACT

Recently, the oscillating magnetocaloric effect (OMCE) was proposed and theoretical verified as an intrinsic property of diamagnetic materials; and then it was further optimized considering a monolayer graphene. OMCE biased by an electric field was also deeply investigated, but Joule heating was a problem to be overcome. The present effort thus goes further and analysis of the OMCE of biased bilayer graphene under an electric and magnetic field is carried out. General expressions for the thermodynamic potentials are derived and then the entropy change and the adiabatic temperature change are investigated. We found that the results for bilayer graphenes are very similar to those for monolayer and, in addition, the Joule heating problem could be overcome, since the gate potential is applied between the two layers.

1. Introduction

The magnetocaloric effect (MCE) is an interesting property in which magnetic materials, under a magnetic field change, are able to exchange heat with a thermal reservoir (considering an isothermal process), or even change its temperature (considering an adiabatic process). This effect is completely analogous to the compression–expansion thermal-mechanical cycle; and therefore the main purpose of the scientific community is to built a thermomagnetic machine, to substitute, in a near future, those standard, non-economical and non-environmental friendly Freon-like refrigerators [1,2]. It is straightforward to see that $\Delta T = T_f - T_i$, obtained from the adiabatic condition $S(T_i, 0) = S(T_f, H)$, characterizes the adiabatic process; while $\Delta S = S(T, H) - S(T, 0)$ characterizes the magnetic entropy change, obtained from the isothermal process.

The magnetic entropy change is larger in magneto-ordered materials in the vicinity of phase transitions [1,3] and therefore the strongest MCE manifests in ferro- and antiferromagnetic materials [1–6]. Due to the above, a deeper evaluation of this effect in diamagnetic materials was only reported recently [7–17]; namely in 3D, 2D and quasi-1D non-relativistic materials [7,8,11] and, in addition, on monolayer graphene (MG) [9,12,13]. The most interesting point of these contributions is that the new oscillatory character of the MCE as a function of the reciprocal magnetic field $1/H$ is analogous to the de Haas-van Alphen effect. Other interesting aspect of the oscillating magnetocaloric effect (OMCE) is its maximum value for the magnetic entropy change even without phase transition (in high contrast to ordered materials that need a phase transition to maximize the MCE). In addition, deeper analysis of the OMCE on graphenes shows that the temperature in which the OMCE peaks are almost 100 times higher than non-relativistic diamagnetic materials (Gold, for instance). The OMCE also provides additional information about the system, like metrology of fundamental physical constants [12]. Thus, OMCE has interesting features that deserve to be further and deeper investigation; and graphenes are materials with the highest potential to show peculiar behaviors, mainly due to its relativistic features. Following this idea, the magnetocaloric properties of graphenes biased by an electric field were deeply investigated [15], but a drawback appeared: Joule heating is quite bigger than the magnetocaloric magnitude. The aim of this effort is therefore to overcome this problem. A straightforward idea is to consider a bilayer graphene with gate voltage applied between those two layers, avoiding the Joule heating.

Indeed, bilayer graphene (BG) is of special interest. From the theoretical viewpoint, BG is attractive due to the fact that electrons are described by an unusual wave equation, that has fourth-order derivatives, in contrast to the Shrödinger and Dirac equations [15]. In addition, an applied gate potential, perpendicular to the carbon planes, opens a gap in the energy spectrum of BG [16]. On the other hand, from the applied point of view, BG and MG-based devices may become promising materials for modern nanoelectronics [15], due to, for instance, an anomalous growth of the thermopower [17], unusual quantum Hall effect [18], exotic electrodynamic effects [19], giant magneto-optical [20] and thermomagnetic [21] effects.

In the present paper, thus, we deeply analyzed the OMCE in a BG and described the physical mechanisms that rule the oscillatory behavior of the magnetocaloric potentials as a function of

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temperature, reciprocal magnetic field and gate potential. We then found on BG a solution to observe a genuine OMCE, biased by an electric field, and without the inconvenience of Joule heating.

2. Bilayer graphene: electronic spectrum and Landau levels

For this effort, we will consider only the simplest theory of electronic spectrum and Landau levels – but, as will be clear further in the text, this procedure is enough to obtain the desired OMCE features of BG. The unit cell of BG has four carbon atoms and, consider the so-called AB stacking, the effective Hamiltonian can be written as [15]

$$\mathcal{H}=\begin{pmatrix} U & t_1^* & t_\perp & 0 \\ t_1 & U & 0 & 0 \\ t_\perp & 0 & -U & t_1 \\ 0 & 0 & t_1^* & -U \end{pmatrix} \tag{1}$$

where

$$\frac{t_1}{t}=e^{iq_x a}+e^{-iq_x a/2}e^{iq_y\sqrt{3}a/2}+e^{-iq_x a/2}e^{-iq_y\sqrt{3}a/2} \tag{2}$$

and t is the nearest-neighbor hopping energy (hopping between different sub-lattices in a monolayer). In addition, q represents the two-dimensional wave vector of electrons, a is the carbon–carbon distance in a monolayer, $t_\perp$ is the hopping energy between two layers, i.e. hopping energy between A_1 and A_2 atoms (A_1 and B_1 are sub-lattices of the first layer, while A_2 and B_2 are sub-lattices of the second layer), and, finally, U is the gate potential, that, on its turn, tune the gap. Acceptable values for t and $t_\perp$ are shown in Table 1, and, for the present effort, we will consider $t_\perp=0.4$ eV and $t=2.8$ eV.

The next step is to consider the region of momentum space in the vicinity of Dirac point and, in this region, the energy spectrum of the MG has a simple form:

$$\frac{t_1}{t}=\pm\frac{3}{2}a|\vec{k}| \tag{3}$$

where $\vec{k}=\vec{q}-\vec{Q}$ and $\vec{Q}=(2\pi/3a, 2\pi/3\sqrt{3}a)$ is the wave vector corresponding to the Dirac point. Above, $\pm$ represents conduction ('+') and valence ('−') bands. Thus, the Hamiltonian on Eq. (1) gives the energy spectrum:

$$\epsilon(U,k)=\nu_b\left\{v_F^2h^2k^2+\frac{t_\perp^2}{2}+U^2 +\nu_{sb}\left[(t_\perp^2+4U^2)v_F^2h^2k^2+\frac{t_\perp^4}{4}\right]^{1/2}\right\}^{1/2} \tag{4}$$

where $v_F=3ta/2h=10^8$ cm/s is the Fermi velocity of electrons; $\nu_b=\pm1$ is a band index ($\nu_b=+1$ corresponds to conduction band and $\nu_b=-1$ corresponds to valence band); $\nu_{sb}=\pm1$ is a sub-band index ($\nu_{sb}=+1$ corresponds to the sub-band I and $\nu_{sb}=-1$ corresponds to the sub-band II). Application of a voltage opens a gap

$$\Delta=\left[\frac{t_\perp^2U^2}{t_\perp^2+U^2}\right]^{1/2} \tag{5}$$

Table 1
Values of hopping energies.

Reference	t (eV)	$t_\perp$ (eV)
Experiment [22] (IR spec.)	–	0.378(5)
Experiment [23] (IR spec.)	3.2(3)	0.381(3)
Experiment [24] (IR spec.)	3.0	0.40
Experiment [25] (Raman)	2.9	0.30
Experiment [26] (Raman)	3.0	0.35
Theory [27] (ab initio)	2.6	0.34
Theory [28] (ab initio)	3.4013	0.3963

in the energy spectrum, as can be seen in Fig. 1; and it is reasonable, since an external electric field breaks the symmetry between first and second layers.

To study the magnetic oscillations, we considered the quantization rule in the Lifshitz–Onsager form [29], since it has been successfully used in recent papers [30–32]. For a 2D electrons gas, the area enclosed by an electron trajectory in the momentum space is [29,33]

$$A(\epsilon)=\frac{2\pi heH}{c}(n+\gamma_\sigma) \tag{6}$$

where $n=0,1,2,\ldots$,

$$\gamma_\sigma=\gamma+\frac{m(\mu)}{2m}\sigma, \tag{7}$$

$$m(\epsilon)=\frac{1}{2\pi}\frac{dA(\epsilon)}{d\epsilon} \tag{8}$$

is the electron cyclotron mass [33], m is the electron mass, $\sigma=\pm1$ and, finally, $\gamma=1/2$ for non-relativistic gas and $\gamma=0$ for graphenes. In the present effort, the Zeeman splitting of the Landau levels is ignored, i.e., we assume that $m(\mu)/m=0$ [34]. Considering $A(\epsilon)=\pi R^2(\epsilon)$, where $R(\epsilon)$ is the radius of an electron trajectory with constant energy, we can write therefore:

$$A(\epsilon)=\frac{\pi}{v_F^2}\left\{\epsilon^2+U^2+[(4U^2+t_\perp^2)\epsilon^2-U^2t_\perp^2]^{1/2}\right\} \tag{9}$$

Finally, the Landau levels can be derived from Eqs. (4) and (6); and reads then as

$$\epsilon_n=\left\{\frac{2heHnv_F^2}{c}+\frac{t_\perp^2}{2}+U^2 -\left[\left(t_\perp^2+4U^2\right)\frac{2heHnv_F^2}{c}+\frac{t_\perp^4}{4}\right]^{1/2}\right\}^{1/2} \tag{10}$$

It is important to stress that the limit in which $t_\perp$ and U tends to zero, Eq. (9) reads as $A(\epsilon)=\pi\epsilon^2/v_F^2$ and Eq. (10) reads as $\epsilon_n=\sqrt{2heHn/c}$. Note these are the results for monolayer graphene [15] and the present evaluation can successful recover those.

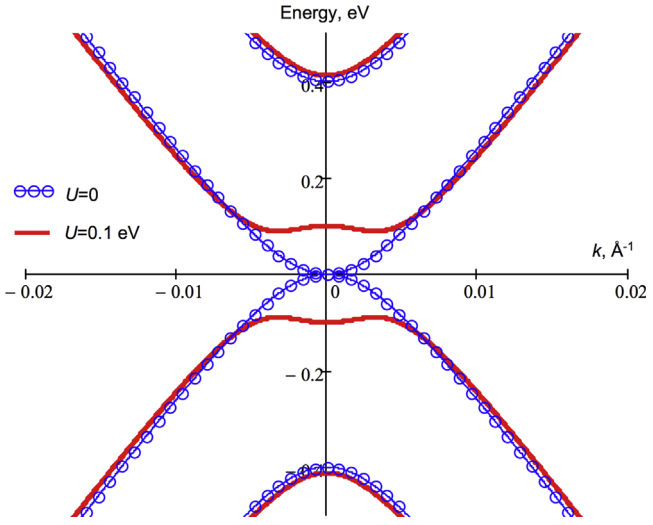

Fig. 1. (Color online) Energy spectrum of a bilayer graphene, where U is the gate potential between these two layers.